

[JP,09-097676,A(1997)]

Japanese (PDF)

File Wrapper Information

FULL CONTENTS CLAIM + DETAILED DESCRIPTION
TECHNICAL FIELD PRIOR ART EFFECT OF THE
INVENTION TECHNICAL PROBLEM MEANS EXAMPLE

[Translation done.]

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Notes:

1. Untranslatable words are replaced with asterisks (* **).
2. Texts in the figures are not translated and shown as it is.

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Dictionary: Last updated 05/30/2008 / Priority:

FULL CONTENTS**[Claim(s)]**

[Claim 1] In the organic electroluminescence element equipped with the organic compound thin film layer containing a luminous layer or a luminous layer between an anode and the negative pole One sort of metallic elements (A) as which the negative pole is chosen from among the alkaline metal which is less than 4.0eV of work functions, alkaline-earth metals, or a rare earth element, One sort of metallic elements (B) chosen from the thing of 4.0eV or more of work functions, The organic electroluminescence element characterized by being the metallic elements which were not chosen above and being the 3 yuan alloy which consists of one sort of metallic elements (C) chosen from Y, Gd, Nd, Sc, La, Zn, Mg, Zr, Cd, In, Mn, Pb, V, Ga, Ti, Bi, Sn, and Cr.

[Claim 2] The organic electroluminescence element according to claim 1 which are one sort of metallic elements as which the metallic elements (B) chosen as a thing of 4.0eV or more of

[Translation done.]

work functions are chosen from In, Pb, Ag, aluminum, Bi, Sn, Sb, Cu, Co, Au, nickel, Pd, and Pt.

[Claim 3] The organic electroluminescence element according to claim 1 or 2 whose rate in the inside of a 3 yuan alloy is 0.1-50at. % although chosen as metallic elements (C).

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the organic electroluminescence (EL) element used for the source of flat light, or a display, and relates to a long lasting light emitting element especially with little luminescence degradation.

[0002]

[Description of the Prior Art] Promising ** of the use as an inexpensive solid luminescence type large area full color display element is carried out, and, as for EL element which uses an organic substance, many development is performed. Generally EL consists of a pair of opposite electrodes which sandwiched the luminous layer and this layer. When **** is impressed between two electrodes, an electron is poured in from the negative pole side and, as for luminescence, an electron hole is poured in from the anode side. Furthermore, when this electron re-combines with an electron hole in a luminous layer and an energy level returns from a conducting zone to a valence band, it is the phenomenon which releases energy as a light.

[0003] The conventional organic EL device had high drive voltage compared with inorganic EL element, and luminescence luminosity and its luminous efficiency were also low. Moreover, characteristic degradation had not resulted in utilization remarkably, either. The organic EL device which laminated the thin film containing an organic compound with the high fluorescence quantum efficiency which emits light by the low voltage not more than 10V is reported, and the interest is attracted in recent years (refer to it applied FIJIKUSU Letters, 51 volumes, 913 pages, and 1987). By this method, the metal chelate complex was used for the phosphor layer, the amine compound was used for the hole injection layer, high-intensity green luminescence has been obtained, 1000cd/m² and the maximum luminous efficiency attain 1.5 lm/W on the direct-current voltage of 6-7V, and luminosity has the performance near a practical use field. However, there is a big problem that the organic EL device by the present is inferior to the stability at the time of repetition use and long-term preservation in the atmosphere although luminescence intensity is being improved

by improvement of composition.

[0004] As one of the means which improves luminescence luminosity and efficiency, in order to raise electronic pouring efficiency, using the alloy containing a small metal and small it of a work function as the negative pole is performed. For example, the example to which the example which mainly used Mg alloy used Li alloy for JP,H4-212287,A, JP,H5-121172,A, JP,H5-159882,A, and JP,H5-198380,A is indicated by JP,H2-15595,A. However, since a small metal of a work function, i.e., the metal which oxidizes easily, is included by these methods, Degradation of the negative pole progresses by moisture and oxygen in the atmosphere, and also when the rise of lower luminance or drive voltage, and the formation and increase of a nonluminescent portion which are further called a dark spot neglect it under the atmosphere, without [under a drive with the passage of time or] making light emit, they are observed.

[0005]

[Problem(s) to be Solved by the Invention] The purpose of this invention is in offer of an organic EL device which was excellent in the stability in the time of repetition use, maintaining the initial characteristics of an element, such as luminescence luminosity and drive voltage. As a result of this invention persons' inquiring wholeheartedly, the organic EL device which uses the 3 yuan alloy which consists of combination of a specific metal with which work functions differ for the negative pole finds out passing and excelling in the Tokiyasu quality, and came to make this invention.

[0006]

[Means for Solving the Problem] That is, this invention is the organic electroluminescence element which equipped the thing about the organic electroluminescence element characterized by being an alloy of 3 yuan with the organic compound thin film layer which is characterized by providing the following, and which contains a luminous layer or a luminous layer between an anode and the negative pole. One sort of metallic elements (A) as which the negative pole is chosen from among the alkaline metal which is less than 4.0eV of work functions, alkaline-earth metals, or a rare earth element One sort of metallic elements (B) chosen from the thing of 4.0eV or more of work functions One sort of metallic elements (C) which are the metallic elements which were not chosen above and chosen from Y, Gd, Nd, Sc, La, Zn, Mg, Zr, Cd, In, Mn, Pb, V, Ga, Ti, Bi, Sn, and Cr [0007] Here, a work function differs in a value greatly according to metallic elements, the form of other material, or a measuring method and conditions in many cases, and differs also in the

value reported with literature. In this invention, the work function of the main metallic elements adopted the value indicated to 493 pages of the basic volume II chemistry manual revised [the 3rd edition (edited by Chemical Society of Japan) of]. Although some are greatly different from the value indicated to JP,H2-15595,A in these, it is not limited to the classification of the metallic elements in this invention, and selection by these literature values.

[0008] It is the fall of the drive voltage by (A) mainly raising electronic pouring efficiency like the conventional 2 yuan alloy as for the metallic elements (A) in the negative pole, and the role of (B), and improvement in luminous efficiency, and (B) is mainly stabilization of (A), and improvement in the adhesion nature to an organic layer. (C) added to this as the 3rd ingredient stabilizes (A) chemically. Maintain the work function of (A) or reduce the work function as an alloy further. The precise nature of an alloy is raised or it has 1 of the effects of preventing osmosis of moisture inside an alloy, or oxygen by controlling oxidization of an alloy or making the oxide layer of (C) from equalizing alloy composition on the surface, or two or more. Although it depends also on the kind of (C), which effect is [among these] discovered may change with kinds of (A) and (B), even if it is the same (C). In [any] an effect, it contributes to (C) making the temporal stability of the negative pole increase harnessing the character of the negative pole, and the good characteristic of (A) especially finally.

[0009] As metallic elements (A) which are less than 4.0eV of work functions in this invention Specifically Cs (1.95), Rb (2.16), K (2.28), Alkaline metals, such as Na (2.36) and Li (2.93), Ba (2.52), There are rare earth elements, such as alkaline-earth metals, such as Sr, Ca (2.9), and Mg (3.66), Eu (2.5), Yb (2.6), Sm (2.7), Ce (2.9), Er (2.97), Gd (3.1), Y (3.1), Nd (3.2), La (3.5), and Sc (3.5). Here, the value in a parenthesis is indicated in the above-mentioned chemistry manual (unit: eV). About Sr, there was no description in this literature.

[0010] In this invention although, as for metallic elements (B) which are 4.0eV or more of work functions, there are varieties In (4.09), Pb (4.25) which are type metallic elements also in this, Ag (4.26), Cu (4.65), Co (5.0), Au (5.1), nickel (5.15), Pd (5.55), and Pt (5.64) which are aluminum (4.28), Bi (4.34), Sn (4.42), Sb (4.55), and a transition metal element are desirable.

[0011] Y (3.1) which is a rare earth element as metallic elements (C) in this invention, Gd (3.1), Nd (3.2), Sc (3.5), La (3.5), Zn (3.63), Mg (3.66), Cd (4.08) which are type metallic elements, Although it is In (4.09), Pb (4.25), Ga (4.3), Bi (4.34), Sn (4.42),

Zr (4.05) that is a transition metal element, Mn (4.1), V (4.3), Ti (4.33), and Cr (4.5) The work function of these metallic elements is in the 3.0eV or more range of 4.5eV or less as it was shown in the parenthesis. Although the metallic elements which can be chosen as (C) can be chosen also as metallic elements (A) or (B) here It is not decided by the combination of the metallic elements in a 3 yuan alloy, and the role of each metallic elements in the inside of an alloy whether it is chosen as metallic elements of which group, and it is not necessarily limited from the beginning. Moreover, there is not necessarily necessity that the value of the work function of metallic elements (C) of 3 yuan is between metallic elements (A) and the value of the work function of (B) in the combination of the metallic elements in an alloy, and a numerical inversion may be between (A), (C), or (B) and (C).

[0012] In this invention, although chosen out of metallic elements (C), as for the rate in the inside of a 3 yuan alloy, it is desirable that it is 0.1-50at.%. at.% is the percentage in an atomic ratio here. If exertion of the effect which contains (C) as it is less than 0.1% cannot be expected but 50% is exceeded, the further improvement in an effect cannot be expected above and other characteristics may be reduced remarkably.

[Embodiment of the Invention]

[0013] an organic EL device -- between an anode and the negative pole -- much more -- or it is the element in which the multilayer organic thin film was formed. In the case of the model, the luminous layer is further prepared between an anode and the negative pole. A luminous layer contains a luminescent material, and in order to make the electron poured in from the electron hole which was poured in from the anode in addition to it, or the negative pole convey to a luminescent material, it may contain electron hole transportation material or electronic transportation material. A multilayer type has the organic EL device laminated with the multilayer composition of (an anode / hole injection layer / luminous layer / negative pole), (an anode / luminous layer / electronic injection layer / negative pole), and the (an anode / hole injection layer / luminous layer / electronic injection layer / negative pole).

[0014] By making it multilayer structure, it becomes possible to make re-combination in the luminous layer of an electron hole and an electron cause efficiently, and the organic EL device can prevent the fall of the luminosity by quenching, or a life further. Moreover, if there is necessity, it can also be used, combining a luminescent material, doping material, the electron hole transportation material that performs carrier transportation, and

electronic transportation material two or more kinds. Moreover, a hole injection layer, a luminous layer, and an electronic injection layer may be formed of the layer composition of two or more layers, respectively, an electron hole or an electron is efficiently poured in from an electrode, and the element structure conveyed in a layer is chosen.

[0015] [a conductive material used for the anode of an organic EL device] A thing with a bigger work function than 4.0eV is suitable, and Carbon, aluminum, Although organic conductivity resin, such as metal oxide, such as tin oxide used for those alloys, such as vanadium, iron, Cobalt, nickel, tungsten, silver, gold, platinum, and palladium, an ITO board, and a NESA board and indium oxide, and also poly CHIOFEN, and polypyrrole, is used It is not limited to these. As long as an anode has necessity, it may be formed of the layer composition of two or more layers.

[0016] In order to make light emit efficiently in an organic EL device, in the luminescence wavelength field of an element, the transparent enough thing of at least one side is desirable. Moreover, it is desirable for a substrate to be also transparent. The above-mentioned conductive material is used for a transparent electrode, and it sets it up to secure predetermined light transmission nature by methods, such as vapor deposition and sputtering. As for the electrode of a luminescence side, it is desirable to make optical transmittance 10% or more. If a substrate has mechanical and thermal intensity, it is not limited if it is transparent, but it is illustrated, transparency resin, such as a glass substrate, a polyethylene board, a poly ether SARUFON board, and a polypropylene board, will be raised.

[0017] The formation of the organic each layer of the organic EL device concerning this invention can apply which method of the wet forming-membranes methods, such as the dry type forming-membranes methods, such as vacuum deposition and sputtering, and spin coating, dipping. Although film thickness in particular is not limited, it needs to set each layer as suitable film thickness. If film thickness is too thick, in order to obtain fixed optical power, big impressed electromotive force will be needed and efficiency will worsen. If film thickness is too thin, even if a pinhole etc. occurs and it impresses **** into a thin film, sufficient luminescence luminosity will not be obtained. The usual film thickness has the still more desirable range of 0.2 micrometer from 10nm, although the range of 10 micrometers is suitable from 5nm.

[0018] The solvent may be any, although the material which forms each layer is dissolved or distributed to suitable solvents, such as chloroform, a tetrahydro franc, and JIOKISAN, in the

case of the wet forming-membranes method and a thin film is formed. Moreover, also in which thin film, you may use suitable resin and a suitable additive agent on a membrane formation disposition for pinhole prevention of a film etc. As possible resin of use, polystyrene, polycarbonate, the poly ant rate, Polyester, polyamide, polyurethane, Pori Sour Von, poly methyl methacrylate, Conductive resin, such as photo electroconductive polymer, such as insulating resin, such as polymethyl acrylate and cellulose, poly-N-vinylcarbazole, and polysilane, Pori CHIOFEN, and polypyrrole, can be mentioned. Moreover, an antioxidant, an ultraviolet ray absorbent, a plasticizer, etc. can be mentioned as an additive agent.

[0019] As the luminescent material which can be used for the organic EL device of this invention, or a doping material ANTORASEN, NAFUTAREN, phenan TOREN, PIREN, TETORASEN, KORONEN, KURISEN, full ORESEIN, PERIREN, phtalo PERIREN, Naphthalo PERIREN, PERINON, phtalo PERINON, naphthalo PERINON, Diphenyl butadiene, tetra-phenyl butadiene, Kumarin, oxadiazole, Aldazine, screw benzoKISAZORIN, screw styryl, PIRAJIN, cyclo pen TAJIEN, The Kino Lynne metal complex, an amino KINORIN metal complex, a benzoKINORIN metal complex, Although there are those derivatives, such as IMIN, diphenyl ethylene, vinyl ANTORASEN, JIAMINOKARUBAZORU, Piran, thiopyran, Pori methine, merocyanine, an imidazo RUKIRETO-ized OKISHINOIDO compound, cinchona bark KURIDON, and rubrene, it is not limited to these.

[0020] The compound which has the capability to convey an electron hole, as an electron hole transportation material, has the electron hole pouring effect of having excelled to the luminous layer or the luminescent material, and prevented movement into the electronic injection layer or electronic transportation material of an exciton generated by the luminous layer, and was excellent in thin film organization potency is mentioned. Specifically A phtalo SHIANIN system compound, a naphthalocyanine system compound, A PORUFIRIN system compound, oxadiazole, bird AZORU, IMIDAZORU, Imidazolone, imidazo RUCHION, PIRAZORIN, PIRAZORON, tetrahydro IMIDAZORU, OKISAZORU, oxadiazole, HIDORAZON, reed RUHIDORAZON, Pori ARIRU Alekan, SUCHIRUBEN, butadiene, benzogin type bird phenylamine, styryl amine type bird phenylamine, Gia Min type bird phenylamine, etc., Although there are polymer materials, such as those derivatives and Pori vinyl KARUBAZORU, polysilane, and a conductive polymer, etc., it is not limited to these.

[0021] The compound which has the capability to convey an electron, as an electronic transportation material, has the electronic pouring effect of having excelled to the luminous layer or the luminescent material, and prevented movement into the hole injection layer or electron hole transportation material of an exciton generated by the luminous layer, and was excellent in thin film organization potency is mentioned. For example, although there are a fluorenone, anthra KINOJI methane, diphenoquinone, thiopyran dioxide, oxadiazole, thiadiazole, tetra-ZORU, PERIREN tetracarboxylic acid, FUREORENIRIDEN methane, anthra KINOJI methane, Antron, etc. and those derivatives It is not limited to these. Moreover, an electronic acceptance substance can be added into electron hole transportation material, an electron release nature substance can be added into electronic transportation material, and a feeling of increase can be carried out.

[0022] As a method of producing the negative pole, methods, such as resistance heating vapor deposition, electron beam vapor deposition, direct-current weld slag, RF weld slag, and ion plating, are mentioned. In the case of the metal which the simplicity of equipment also has and evaporates at low temperature comparatively, resistance heating vapor deposition is very suitable. When using the metal which evaporates at high temperature comparatively, electron beam vapor deposition and a sputtering method are more desirable. As the source of vapor deposition, or a sputtering target, according to a method, even if it is a metal simple substance, it does not matter even if it is an alloy. However, since it is very difficult, as for making it evaporate simultaneously by the ratio of a request of each metal from one sort or two kinds of alloys in resistance heating vapor deposition, it is desirable to carry out simultaneous vapor deposition of 3 yuan using three sources of vapor deposition. However, since it is very unstable and the vapor deposition from a simple substance is slightly difficult for especially a small metal of work functions, such as Li, in the air, it is more desirable to evaporate only low work function metal from the alloy of such low work function metal and the metal of the high-boiling point which does not evaporate simultaneously. Moreover, in a sputtering method, since it is difficult for simple substances, such as Li, to create a target, it is more desirable to create an alloy target and to carry out by RF weld slag which can produce composition comparatively near the original alloy target.

[0023] As long as the alloy produced as the negative pole does not need to form the crystal state or intermetallic compound

which were not necessarily put in order and can demonstrate sufficient effect, in the amorphous state called what is called a solid solution is sufficient as it.

[0024] This invention does not bar making it a 4 yuan or more alloy by further already adding the metal ingredient more than a kind, in order to improve the stability or other characteristics of the negative pole further. Moreover, the negative pole may be constituted by the metal or the alloy of two or more layers if needed, and the composition and ingredient ratio may change continuously by the lower part of a layer, and Uwabe.

[0025] In order to stabilize the negative pole further, you may produce a stable metal or a stable alloy on the negative pole physically and chemically that it is hard to react with these which can penetrate neither moisture nor oxygen easily. These metal or alloys may also be chosen from one sort in the metal ingredient which forms the negative pole, or two sorts, and may completely be formed from another metal ingredient. Moreover, you may form still higher insulating oxide and insulating nitriding thing of barrier property to moisture or oxygen so that the negative pole may be covered.

[0026] It is also possible to prepare a protection layer by a closure film, closure resin, etc. on the surface of an element, or to enclose silicon oil etc., and to protect the whole element for the further improvement in the stability over the temperature of the organic EL device obtained by this invention, humidity, atmosphere, etc.

[0027] As mentioned above, in this invention, since the 3 yuan alloy which becomes the negative pole of an organic EL device from the combination of a specific metal with which work functions differ was used, oxidization of the negative pole was prevented and it became possible to make high stability with the passage of time. moreover, since this element is very stable also to heat or current and usable luminescence luminosity was further obtained practical by the low voltage, it was a big problem to the former -- it was able to pass, degradation in the time and luminescence time were able to be raised sharply, and it was able to use for the improvement in a life of an organic EL device.

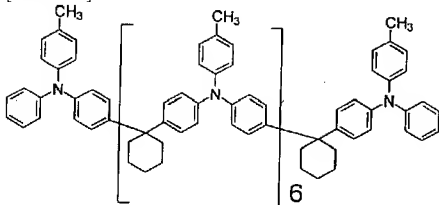
[0028] The organic EL device of this invention can consider application as a flat-panel display and plane photogens, such as a flat TV, to light sources, such as light sources, such as a copying machine and a printer, a liquid crystal display, and instruments, a display board, a beacon light, etc., and the industrial value is very large.

[0029]

[Example] This invention is hereafter explained still in detail based on a work example.

[0030] On the glass board with an ITO electrode washed work-example 1, vacuum deposition of the compound (1) shown by the following chemical structure was carried out, and the hole injection layer of 50nm of film thickness was obtained. Subsequently, the tris (8-hydroxy KINORIN) aluminum complex was made into the luminescent material, vacuum deposition was carried out, and the luminous layer of 50nm of film thickness was created. Moreover, evaporated Mg as metallic elements (A), aluminum was evaporated as metallic elements (B), Zn was simultaneously evaporated from each source of vapor deposition as metallic elements (C), and the negative pole of the film thickness of 200nm of film thickness was formed with the alloy of Mg:aluminum:Zn=10:1:1 by performing vapor codeposition. A hole injection layer, a luminous layer, and the negative pole were vapor-deposited under the conditions of substrate temperature room temperature in the vacuum of 10⁻⁶Torr. Thus, the created organic EL device is the luminescence luminosity of 220cds/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.1 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m² the place which continued the constant current drive -- 500 hours after -- 65cd/m² up to -- **** -- it did not fall. Moreover, most dark spots were not seen. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, the dark spot was not seen but showed uniform luminescence.

[Formula 1]



Compound (1)

[0031] Except using the alloy of composition of Table 1 as the two to work-example 22 negative pole, an organic EL device is created by the same method as a work example 1, and it is

luminescence luminosity [in the direct-current voltage 5V], and initial 100cd/m². The luminosity of 500 hours after when continuing a constant current drive was measured. A result is shown in Table 1. In addition, when Li was vapor-deposited and Sm was vapor-deposited for the alloy of Li:aluminum=1:1, the alloy of Sm:Co=1:5 was used as a source of vapor deposition, and only Li or Sm was evaporated.

[0032]

[Table 1]

実施例	合金組成および比率 A : B : C	5 Vでの初期輝度 (cd/m ²)	500時間後の輝度 (初期100cd/m ²)
2	Mg Al Mn 10 : 1 : 1	200	70
3	Mg Ag Zn 9 : 1 : 2	250	60
4	Mg Ag In 17 : 1 : 2	280	65
5	Li Al Mg 3 : 1 : 6	310	60
6	Li Al In 4 : 2 : 4	320	55
7	Li In Zn 1 : 6 : 3	300	60
8	Sm Al Mn 4 : 5 : 1	270	55
9	Li Al Cr 2 : 90 : 8	210	75
10	Li In Sn 1 : 80 : 19	180	80
11	Yb Al Zn 5 : 2 : 3	290	70
12	Li Sb La 15 : 50 : 35	210	85

[0033]

実施例	合金組成および比率 A : B : C			5 Vでの初期輝度 (cd/m ²)	500時間後の輝度 (初期100cd/m ²)
13	Ca	Bi	Cd		
	20	78	2	330	80
14	Mg	Pb	In		
	4	1	5	280	55
15	Sm	Co	Gd		
	15	84	1	230	65
16	Ca	Sn	Nd		
	40	55	5	240	60
17	Yb	Pt	Pb		
	3	2	5	190	85
18	Mg	Ag	Bi		
	8	1	1	210	70
19	Mg	Cu	Sc		
	7	1	2	180	75
20	Sm	Pd	Ga		
	4	2	4	250	65
21	Mg	Ni	V		
	6	2	2	170	60
22	Mg	Au	Sn		
	8	1	1	220	70

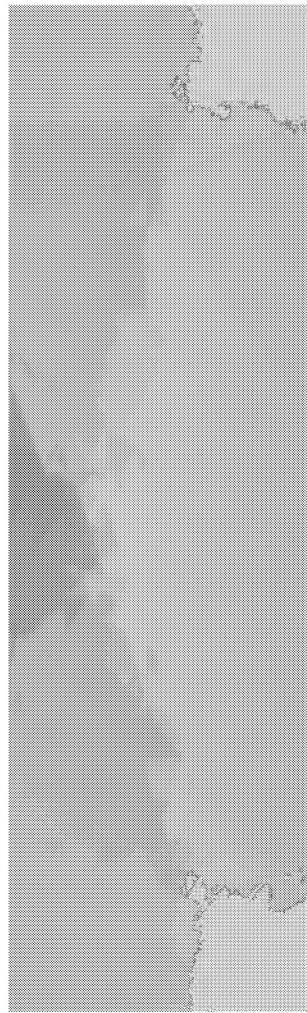
[0034] On the glass board with an ITO electrode washed work-example 23, they are N, N'-(4-methylphenyl)-N, and N'-(4-n-buthylphenyl)- 50nm vacuum deposition was carried out to the order of phenan Trenn 9, 10-Gia Min, and a tris (8-hydroxy KINORIN) aluminum complex, respectively. The electrode of 200nm of film thickness was formed with the alloy moreover mixed by Mg:aluminum:Zn=7:1:2, and the organic EL device was obtained. Each layer was vapor-deposited under the conditions of substrate temperature room temperature in the vacuum of 10-6Torr. This element is 300cd/m2 at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 1.9 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m2 the place which continued the constant current drive -- 500 hours after -- 60cd/m2 up to -- **** -- it did not fall. Moreover, most dark spots were not seen. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, most dark spots were not seen.

[0035] On the glass board with an ITO electrode washed work-example 24, chloroform was made to carry out dissolution

distribution of the compound (1), and the hole injection layer of 50nm of film thickness was obtained by spin coating. Subsequently, 50nm vacuum deposition of the tris (8-hydroxy KINORIN) aluminum complex was carried out as a luminous layer. The electrode of 200nm of film thickness was formed with the alloy moreover mixed by Mg:Ag:Mn=9:1:1, and the organic EL device was obtained. A luminous layer and the negative pole were vapor-deposited under the conditions of substrate temperature room temperature in the vacuum of 10-6Torr. This element is 280cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.4 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m² the place which continued the constant current drive -- 500 hours after -- 75cd/m² up to -- **** -- it did not fall. Moreover, most dark spots were not seen. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, the dark spot was not seen but showed uniform luminescence.

[0036] On the glass board with an ITO electrode washed work-example 25, the hole injection layer and the luminous layer were formed like the work example 24. The electrode of 200nm of film thickness was formed with the alloy moreover mixed by Li:In:Zn=3:3:4, and the organic EL device was obtained. A luminous layer and the negative pole were vapor-deposited under the conditions of substrate temperature room temperature in the vacuum of 10-6 Torr. This element is 200cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.5 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m² the place which continued the constant current drive -- 500 hours after -- 70cd/m² up to -- **** -- it did not fall. Moreover, most dark spots were not seen. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, the dark spot was not seen at all but showed uniform luminescence.

[0037] On the glass board with an ITO electrode washed work-example 26, the hole injection layer and the luminous layer were formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10-6Torr like the work example 1. Using the target moreover divided by the surface ratio of Mg:Ag:Zn=13:1:2, the electrode of the alloy thin film of 200nm of film thickness was formed with the 0.5kW output under Ar gas by RF sputtering method, and the organic EL device was obtained. This element is 230cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained

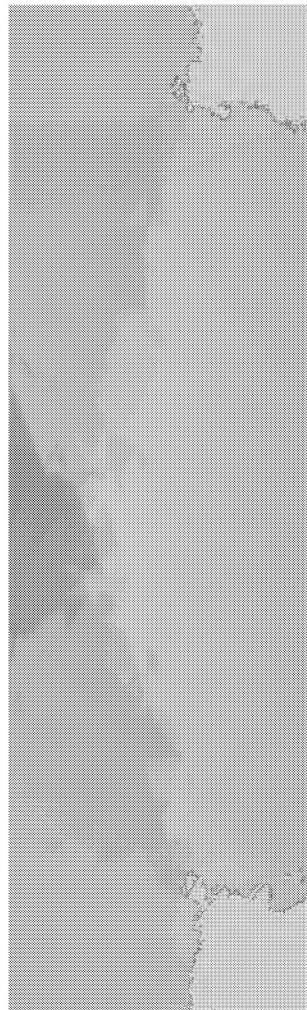


and luminous efficiency was 2.4 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m2 the place which continued the constant current drive -- 500 hours after -- 70cd/m2 up to -- **** -- it did not fall. Moreover, most dark spots were not seen. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, the dark spot was not seen but showed uniform luminescence.

[0038] On the glass board with an ITO electrode washed work-example 27, the hole injection layer and the luminous layer were formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10-6Torr like the work example 1. Using the alloy target moreover beforehand formed by Li:aluminum:Y=5:85:10, the electrode of the alloy thin film of 200nm of film thickness was formed with the 0.5kW output under Ar gas by RF sputtering method, and the organic EL device was obtained. This element is 220cd/m2 at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.3 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m2 the place which continued the constant current drive -- 500 hours after -- 80cd/m2 up to -- **** -- it did not fall. Moreover, a dark spot was not seen, either. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, the dark spot was not seen at all but showed uniform luminescence.

[0039] On the glass board with an ITO electrode washed work-example 28, the hole injection layer and the luminous layer were formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10-6Torr like the work example 1. Using the alloy target moreover beforehand formed by Li:nickel:Zr=1:6:3, the electrode of the alloy thin film of 200nm of film thickness was formed with the 0.5kW output under Ar gas by RF sputtering method, and the organic EL device was obtained. This element is 180cd/m2 at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.5 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m2 the place which continued the constant current drive -- 500 hours after -- 85cd/m2 up to -- **** -- it did not fall. Moreover, a dark spot was not seen, either. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, the dark spot was not seen at all but showed uniform luminescence.

[0040] On the glass board with an ITO electrode washed work-example 29, the hole injection layer and the luminous layer were formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10-6Torr like



the work example 1. Using the alloy target moreover beforehand formed by Li:aluminum:Ti=3:70:27, the electrode of the alloy thin film of 200nm of film thickness was formed with the 0.5kW output under Ar gas by RF sputtering method, and the organic EL device was obtained. This element is 200cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.2 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m² the place which continued the constant current drive -- 500 hours after -- 80cd/m² up to -- **** -- it did not fall. Moreover, a dark spot was not seen, either. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, the dark spot was not seen at all but showed uniform luminescence.

[0041] On the glass board with an ITO electrode washed comparative example 1, the hole injection layer and the luminous layer were formed like the work example 1. The electrode of 200nm of film thickness was formed with the alloy moreover mixed by Mg:In=10:1, and the organic EL device was obtained. Each layer was formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10-6Torr. This element is 230cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.2 lm/W. It is initial luminosity 100cd/m², without closing this element in particular. When a constant current drive is continued, in 500 hours, 70% or more of a luminescence side is covered with a dark spot, and luminosity is also 10cd/m². It has fallen to the following. Moreover, under the atmosphere, after 1000-hour neglect, when light was made to emit, light was hardly emitted.

[0042] On the glass board with an ITO electrode washed comparative example 2, the hole injection layer and the luminous layer were formed like the work example 1. The electrode of 200nm of film thickness was formed with the alloy moreover mixed by Mg:aluminum=8:2, and the organic EL device was obtained. Each layer was formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10-6Torr. This element is 190cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.1 lm/W. the ** which does not close this element in particular -- initial luminosity 100cd/m² when a constant current drive is continued, in 500 hours, 50% or more of a luminescence side is covered with a dark spot -- luminosity -- 15cd/m² up to -- it has fallen. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, it was covered with the dark spot

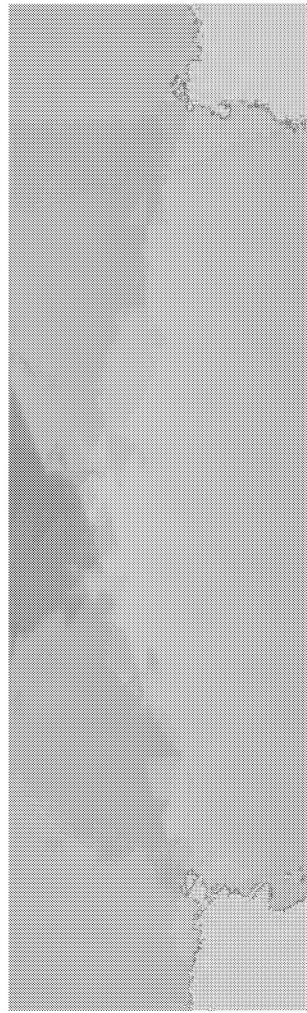


and luminescence was very weak.

[0043] On the glass board with an ITO electrode washed comparative example 3, the hole injection layer and the luminous layer were formed like the work example 1. The electrode of 200nm of film thickness was formed with the alloy moreover mixed by Li:aluminum=3:7, and the organic EL device was obtained. Each layer was formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10⁻⁶Torr. This element is 220cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.7 lm/W. It is initial luminosity 100cd/m², without closing this element in particular. When the constant current drive was continued, in 500 hours, luminosity is 10cd/m². It has fallen to below. A dark spot and luminosity nonuniformity had arisen in the luminescence side. Under the atmosphere, after 1000-hour neglect, when you tried to make it emit light, light was not emitted at all.

[0044] On the glass board with an ITO electrode washed comparative example 4, the hole injection layer and the luminous layer were formed like the work example 24. The electrode of 200nm of film thickness was formed with the alloy moreover mixed by Mg:Ag=10:1, and the organic EL device was obtained. Each layer was formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10⁻⁶Torr. This element is 270cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.5 lm/W. It is initial luminosity 100cd/m², without closing this element in particular. When a constant current drive is continued, in 500 hours, 70% or more of a luminescence side is covered with a dark spot, and luminosity is also 10cd/m². It has fallen to the following. Moreover, under the atmosphere, after 1000-hour neglect, when you tried to make it emit light, light was not emitted at all.

[0045] On the glass board with an ITO electrode washed comparative example 5, the hole injection layer and the luminous layer were formed by vacuum deposition under the conditions of substrate temperature room temperature in the vacuum of 10⁻⁶Torr like the work example 1. Using the alloy target moreover beforehand formed by Mg:Ag=10:1, the electrode of 200nm of film thickness was formed with the 0.5kW output under Ar gas by RF sputtering method, and the organic EL device was obtained. This element is 280cd/m² at the direct-current voltage 5V. Yellowish green luminescence was obtained and luminous efficiency was 2.4 lm/W. the **



which does not close this element in particular -- initial luminosity 100cd/m2 when a constant current drive is continued, in 500 hours, 50% or more of a luminescence side is covered with a dark spot -- luminosity -- 20cd/m2 up to -- it has fallen. Moreover, when light was made to emit after 1000-hour neglect under the atmosphere, it was covered with the dark spot and luminescence was very weak.

[0046] Thus, the organic EL device by a Prior art is initial 100cd/m2. It is 20cd/m2 by the continuous drive of 500 hours. It falls to below, and a luminescence side is covered by a dark spot, or nonuniformity arises. Receiving, the element of this invention is 50cd/m2. The above luminosity was maintained and there was also almost no generating of a dark spot. Moreover, the element of this invention showed uniform luminescence of sufficient luminosity to the organic EL device by a Prior art stopping almost emitting light by neglecting it under the atmosphere. The organic EL device which uses the 3 yuan alloy shown in the negative pole by this invention from the above result attained the reinforcement in a drive and preservation of an element, without reducing luminescence luminosity and luminous efficiency.

[0047] the increase of the luminescent material used attaining reinforcement and combining it the organic EL device of this invention maintaining luminous efficiency and luminescence luminosity, doping material, electron hole transportation material, and electronic transportation material -- admiration -- an agent -- the element production methods, such as resin and electrode material, are not limited.

[0048]

[Effect of the Invention] Since degradation of the negative pole was controlled, compared with the former, the stability with the passage of time over environment of the organic EL device of this invention was high, and it was able to obtain the long lasting organic EL device. It became possible to produce easily a long lasting organic EL device with little degradation with the passage of time, maintaining high luminescence luminosity and high luminous efficiency by using the 3 yuan alloy shown by this invention as the negative pole of an organic EL device by the above.

[Translation done.]

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